PNAS
www.pnas.org

Supplementary Information for

Moiré magnets

Kasra Hejazi, Zhu-Xi Luo and Leon Balents

Leon Balents
E-mail: balents@kitp.ucsb.edu

This PDF file includes:

www.pnas.org/cgi/doi/10.1073/pnas.2000347117

Supporting Information Text

A. Linear wave analysis with anisotropy. In this section, we present the magnon spectrum when the anisotropy is present, and separately discuss the cases of Ising anisotropy ($\beta > 0$) and XY anisotropy ($\beta < 0$).

Ising Anisotropy. Following the parametrization in equation (15), The Hamiltonian near the saddle point $\mathbf{N}_l^{\rm cl}$ is

$$\begin{aligned}\mathsf{H} =&(\nabla_{\mathsf{x}}\phi_1)^2 + (\nabla_{\mathsf{x}}\phi_2)^2 - \alpha\hat{\Phi}(\mathsf{x})\cos(\phi_1-\phi_2) - \beta(\cos^2\phi_1 + \cos^2\phi_2)\\ &+(\nabla_{\mathsf{x}}u_1)^2 + (\nabla_{\mathsf{x}}u_2)^2 + (\nabla_{\mathsf{x}}v_1)^2 + (\nabla_{\mathsf{x}}v_2)^2 - v_1^2(\nabla_{\mathsf{x}}\phi_1)^2 - v_2^2(\nabla_{\mathsf{x}}\phi_2)^2\\ &-\frac{1}{2}\alpha\hat{\Phi}(\mathsf{x})\left[2v_1v_2 + \cos(\phi_1-\phi_2)(2u_1u_2 - u_1^2 - u_2^2 - v_1^2 - v_2^2)\right]\\ &+\beta(u_1^2\cos 2\phi_1 + u_2^2\cos 2\phi_2 + v_1^2\cos^2\phi_1 + v_2^2\cos^2\phi_2) + \ldots,\end{aligned} \tag{1}$$

where the first line is the 0th order contribution, and the dots in the end represent higher-order terms in u and v. Switching to the symmetric and anti-symmetric basis, this becomes

$$\begin{aligned}\mathsf{H} =&\mathsf{H}_{cl} + \frac{1}{2}\left[(\nabla_{\mathsf{x}}u_s)^2 + (\nabla_{\mathsf{x}}u_a)^2 + (\nabla_{\mathsf{x}}v_s)^2 + (\nabla_{\mathsf{x}}v_a)^2\right]\\ &-\frac{1}{8}\left\{(v_a^2+v_s^2)[(\nabla_{\mathsf{x}}\phi_a)^2 + (\nabla_{\mathsf{x}}\phi_s)^2] + 4v_sv_a\nabla_{\mathsf{x}}\phi_a\nabla_{\mathsf{x}}\phi_s\right\}\\ &+\frac{\alpha}{4}\hat{\Phi}(\mathsf{x})\left[-v_s^2(1-\cos\phi_a) + v_a^2(1+\cos\phi_a) + 2u_a^2\cos\phi_a\right]\\ &+\frac{\beta}{4}\left[(v_s^2+v_a^2) + (2u_s^2+2u_a^2+v_s^2+v_a^2)\cos\phi_s\cos\phi_a - (4u_su_a + 2v_sv_a)\sin\phi_s\sin\phi_a\right]\end{aligned} \tag{2}$$

where H_{cl} is defined in equation (10) of the main text.

For the second order terms of the Hamiltonian, we go to the Lagrangian by

$$\mathsf{L}_2 = \frac{1}{2v^2}\left(|\partial_t u_s|^2 + |\partial_t u_a|^2 + |\partial_t v_s|^2 + |\partial_t v_a|^2\right) - \mathsf{H}_2, \tag{3}$$

which leads to the following coupled linear wave equations for u, v's:

$$\begin{aligned}\partial_t^2 u_s &= v^2q_m^2\left[\nabla_{\mathsf{x}}^2u_s - \beta(\cos\phi_s\cos\phi_a u_s - \sin\phi_s\sin\phi_a u_a)\right],\\ \partial_t^2 u_a &= v^2q_m^2\left[\nabla_{\mathsf{x}}^2u_a - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a u_a - \beta(\cos\phi_s\cos\phi_a u_a - \sin\phi_s\sin\phi_a u_s)\right],\\ \partial_t^2 v_s &= v^2q_m^2[\nabla_{\mathsf{x}}^2v_s + \frac{1}{4}\left(v_s(\nabla_{\mathsf{x}}\phi_s)^2 + v_s(\nabla_{\mathsf{x}}\phi_a)^2 + 2v_a\nabla_{\mathsf{x}}\phi_s\nabla_{\mathsf{x}}\phi_a\right) + \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1-\cos\phi_a)v_s\\ &\quad - \frac{\beta}{2}(v_s + v_s\cos\phi_s\cos\phi_a - v_a\sin\phi_s\sin\phi_a)],\\ \partial_t^2 v_a &= v^2q_m^2[\nabla_{\mathsf{x}}^2v_a + \frac{1}{4}\left(v_a(\nabla_{\mathsf{x}}\phi_s)^2 + v_a(\nabla_{\mathsf{x}}\phi_a)^2 + 2v_s\nabla_{\mathsf{x}}\phi_s\nabla_{\mathsf{x}}\phi_a\right) - \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1+\cos\phi_a)v_s\\ &\quad - \frac{\beta}{2}(v_a + v_a\cos\phi_s\cos\phi_a - v_s\sin\phi_s\sin\phi_a)].\end{aligned} \tag{4}$$

Taking $u_{s/a}(\mathsf{x},t) = e^{i\omega t}u_{s/a}(\mathsf{x})$ and using the Bloch ansatz (with $\mathsf{k} = \mathbf{k}/q_m$ being the dimensionless quasi-momentum vector),

$$u_{s/a}(\mathsf{x}) = \hat{u}_{s/a}(\mathsf{x})e^{i\mathsf{k}\cdot\mathsf{x}}, \quad v_{s/a}(\mathsf{x}) = \hat{v}_{s/a}(\mathsf{x})e^{i\mathsf{k}\cdot\mathsf{x}}, \tag{5}$$

we obtain

$$\begin{aligned}\omega^2\hat{u}_s &= -v^2q_m^2\left[(\nabla_{\mathsf{x}} + i\mathsf{k})^2\hat{u}_s - \beta(\cos\phi_s\cos\phi_a\hat{u}_s - \sin\phi_s\sin\phi_a\hat{u}_a)\right],\\ \omega^2\hat{u}_a &= -v^2q_m^2\left[(\nabla_{\mathsf{x}} + i\mathsf{k})^2\hat{u}_a - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a\hat{u}_a - \beta(\cos\phi_s\cos\phi_a\hat{u}_a - \sin\phi_s\sin\phi_a\hat{u}_s)\right],\\ \omega^2\hat{v}_s &= -v^2q_m^2[(\nabla_{\mathsf{x}} + i\mathsf{k})^2\hat{v}_s + \frac{1}{4}\left(\hat{v}_s(\nabla_{\mathsf{x}}\phi_s)^2 + \hat{v}_s(\nabla_{\mathsf{x}}\phi_a)^2 + 2\hat{v}_a\nabla_{\mathsf{x}}\phi_s\nabla_{\mathsf{x}}\phi_a\right) + \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1-\cos\phi_a)\hat{v}_s\\ &\quad - \frac{\beta}{2}(\hat{v}_s + \hat{v}_s\cos\phi_s\cos\phi_a - \hat{v}_a\sin\phi_s\sin\phi_a)],\\ \omega^2\hat{v}_a &= -v^2q_m^2[(\nabla_{\mathsf{x}} + i\mathsf{k})^2\hat{v}_a + \frac{1}{4}\left(\hat{v}_a(\nabla_{\mathsf{x}}\phi_s)^2 + \hat{v}_a(\nabla_{\mathsf{x}}\phi_a)^2 + 2\hat{v}_s\nabla_{\mathsf{x}}\phi_s\nabla_{\mathsf{x}}\phi_a\right) - \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1+\cos\phi_a)\hat{v}_a\\ &\quad - \frac{\beta}{2}(\hat{v}_a + \hat{v}_a\cos\phi_s\cos\phi_a - \hat{v}_s\sin\phi_s\sin\phi_a)].\end{aligned} \tag{6}$$

In the collinear phase, the Neel vectors in the two layers are uniform and point either to the $+\hat{z}$ or the $-\hat{z}$ direction. Namely, there are four possible combinations that are degenerate in energy: $(\phi_s,\phi_a) = (0,0)$, $(2\pi,0)$, and (π,π), $(\pi,-\pi)$. The last two of them satisfying $\cos\phi_s = \cos\phi_a = -1$ have the same discrete symmetry as that of the twisted-s phase, i.e. a simultaneous

spin reflection $N_z \to -N_z$ and layer exchange. For this type of solutions, Eq. (6) reduces to a pair of degenerate equations for (u_s, v_a) and (u_a, v_s):

$$\begin{aligned}\omega^2 \hat{u}_s &= -v^2 q_m^2 [(\nabla_{\mathsf{x}} + i\mathsf{k})^2 - \beta]\hat{u}_s, \\ \omega^2 \hat{u}_a &= -v^2 q_m^2 [(\nabla_{\mathsf{x}} + i\mathsf{k})^2 + \alpha\hat{\Phi}(\mathsf{x}) - \beta]\hat{u}_a.\end{aligned} \tag{7}$$

For the other two solutions with $\cos\phi_s = \cos\phi_a = 1$, Eq. (6) reduces to a pair of degenerate equations for (u_s, v_s) and (u_a, v_a) instead. The u_s and u_a modes satisfy the same set of equations as in Eq. (7) upon substituting $\alpha \to -\alpha$. We will choose the $\cos\phi_s = \cos\phi_a = -1$ case below for concreteness.

The twisted-a phase is the only one that involves the interplay between the symmetric and anti-symmetric modes. As can be observed from Eq. (6), the $u_s,\ u_a$ modes are mixed, so are v_s and v_a; the four branches thus combine into two, which we will simply label as u and v. There is one Goldstone mode in the v-branch.

In the twisted-s phase, the four equations decouple, and there is again one Goldstone mode corresponding to the out-of-plane rotation v_s.

$$\begin{aligned}\omega^2 \hat{u}_s &= -v^2 q_m^2 \left[(\nabla_{\mathsf{x}} + i\mathsf{k})^2 + \beta\cos\phi_a\right]\hat{u}_s, \\ \omega^2 \hat{u}_a &= -v^2 q_m^2 \left[(\nabla_{\mathsf{x}} + i\mathsf{k})^2 - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a + \beta\cos\phi_a\right]\hat{u}_a, \\ \omega^2 \hat{v}_s &= -v^2 q_m^2 [(\nabla_{\mathsf{x}} + i\mathsf{k})^2 + \frac{1}{4}(\nabla_{\mathsf{x}}\phi_a)^2 + \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1-\cos\phi_a) - \frac{\beta}{2}(1-\cos\phi_a)]\hat{v}_s, \\ \omega^2 \hat{v}_a &= -v^2 q_m^2 [(\nabla_{\mathsf{x}} + i\mathsf{k})^2 + \frac{1}{4}(\nabla_{\mathsf{x}}\phi_a)^2 - \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1+\cos\phi_a) - \frac{\beta}{2}(1-\cos\phi_a)]\hat{v}_a.\end{aligned} \tag{8}$$

The magnon bands in the three phases are shown in figure S1.

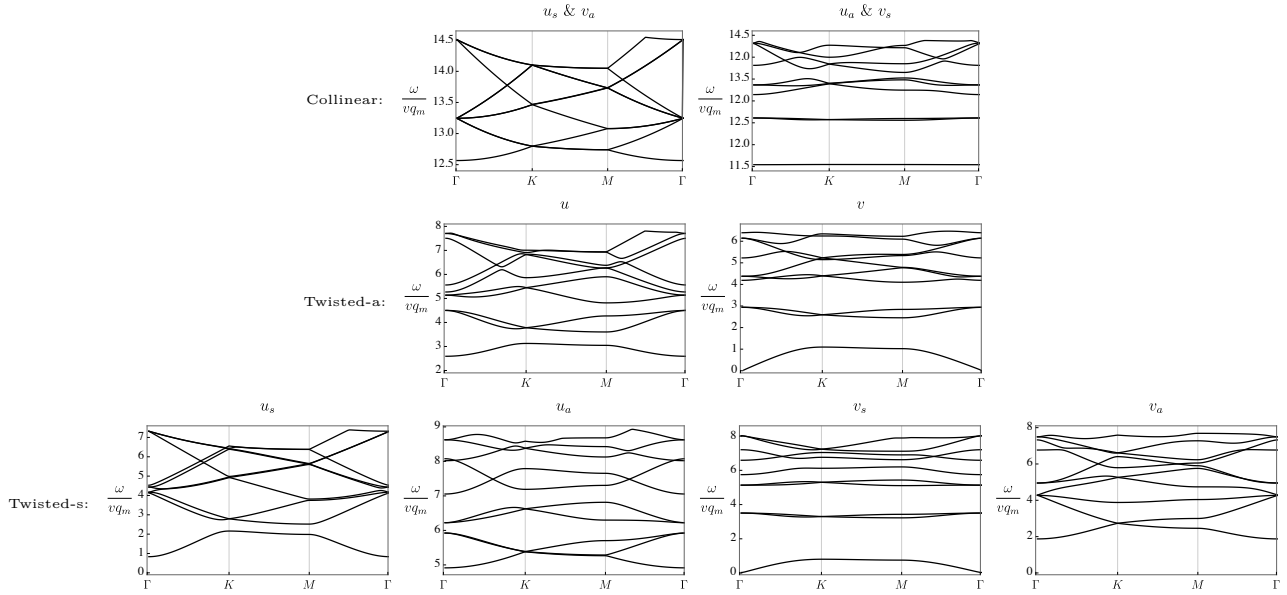

Fig. S1. Top row: The ten lowest magnon bands in the collinear phase for the four branches, where we have chosen $\cos\phi_s = \cos\phi_a = -1$. The dimensionless parameters are $\alpha = 1$, $\beta = 9$. Middle: Magnon bands for the two branches $u,\ v$ in the twisted-a phase at $\alpha = 9$, $\beta = 1$. Bottom: Twisted-s phase for the four decoupled branches at $\alpha = 2$, $\beta = 0.2$.

Similar to the isotropic case, the magnon bands flatten at large α due to their confinement in the disconnected domains in a large potential. Below we show an example $\alpha = 19,\ \beta = 9$ in the twisted-a phase.

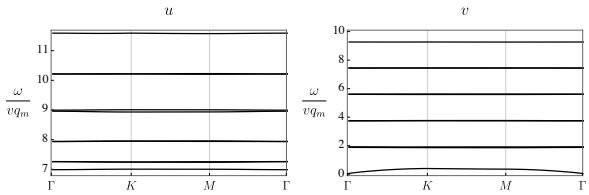

Fig. S2. Flattening of magnon bands in the twisted-a phase at $\alpha = 19,\ \beta = 9$.

XY Anisotropy. Now we turn to the case $d < 0$, where the Neel vectors tend to lie in the XY plane. We thereby choose the following ansatz:

$$\begin{aligned} \mathbf{N}_l &= \sqrt{1-u_l^2-v_l^2}\mathbf{N}_l^{cl}(\mathsf{x}) + u_l\mathbf{u}_l(\mathsf{x}) + v_l\mathbf{v}_l(\mathsf{x}), \\ \mathbf{N}_l^{cl} &= \sin\phi_l\hat{\mathbf{x}} + \cos\phi_l\hat{\mathbf{y}}, \quad \mathbf{u}_l = \cos\phi_l\hat{\mathbf{x}} - \sin\phi_l\hat{\mathbf{y}}, \quad \mathbf{v}_l = \hat{\mathbf{z}}. \end{aligned} \tag{9}$$

The Hamiltonian at the saddle point $\mathbf{N}_l^{cl}$ is $\mathsf{H}_{cl} = \frac{1}{2}\left[(\nabla_\mathsf{x}\phi_s)^2 + (\nabla_\mathsf{x}\phi_a)^2\right] - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a$. Classically, the system behaves as if there is no anisotropy, and ϕ_s is uniform everywhere. Near the saddle point, following the same procedure as the section above, we obtain in the symmetric/anti-symmetric basis:

$$\begin{aligned} \mathsf{H}_2 =& \frac{1}{2}\left[(\nabla_\mathsf{x}u_s)^2 + (\nabla_\mathsf{x}u_a)^2 + (\nabla_\mathsf{x}v_s)^2 + (\nabla_\mathsf{x}v_a)^2\right] - \frac{1}{8}(v_a^2+v_s^2)(\nabla_\mathsf{x}\phi_a)^2 \\ &+ \frac{\alpha}{4}\hat{\Phi}(\mathsf{x})\left[-v_s^2(1-\cos\phi_a) + v_a^2(1+\cos\phi_a) + 2u_a^2\cos\phi_a\right] + \frac{\beta}{2}\left[(v_s^2+v_a^2)\right], \end{aligned} \tag{10}$$

where the parametrization $\beta = 2|d|/\rho q_m^2$ has been used in the last line, which is slightly modified from that in the main text (where $\beta = 2d/\rho q_m^2$). The corresponding Lagrangian then leads to the following linear wave equations:

$$\begin{aligned} &\partial_t^2 u_s = v^2 q_m^2 \nabla_\mathsf{x}^2 u_s, \quad \partial_t^2 u_a = v^2 q_m^2\left[\nabla_\mathsf{x}^2 u_a - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a u_a\right], \\ &\partial_t^2 v_s = v^2 q_m^2[\nabla_\mathsf{x}^2 v_s + \frac{1}{4}v_s(\nabla_\mathsf{x}\phi_a)^2 + \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1-\cos\phi_a)v_s - \beta v_s], \\ &\partial_t^2 v_a = v^2 q_m^2[\nabla_\mathsf{x}^2 v_a + \frac{1}{4}v_a(\nabla_\mathsf{x}\phi_a)^2 - \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1+\cos\phi_a)v_s - \beta v_a]. \end{aligned} \tag{11}$$

All the four branches decouple and almost reduce to the the isotropic form as in the main text, up to the constant shift of β in the v-branches. With the Bloch ansatz, the above equations become

$$\begin{aligned} &\omega^2\hat{u}_s = -v^2q_m^2\left[(\nabla_\mathsf{x} + i\mathsf{k})^2\hat{u}_s\right], \\ &\omega^2\hat{u}_a = -v^2q_m^2\left[(\nabla_\mathsf{x} + i\mathsf{k})^2\hat{u}_a - \alpha\hat{\Phi}(\mathsf{x})\cos\phi_a\hat{u}_a\right], \\ &\omega^2\hat{v}_s = -v^2q_m^2[(\nabla_\mathsf{x} + i\mathsf{k})^2\hat{v}_s + \frac{1}{4}\hat{v}_s(\nabla_\mathsf{x}\phi_a)^2 + \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1-\cos\phi_a)\hat{v}_s - \beta\hat{v}_s], \\ &\omega^2\hat{v}_a = -v^2q_m^2[(\nabla_\mathsf{x} + i\mathsf{k})^2\hat{v}_a + \frac{1}{4}\hat{v}_a(\nabla_\mathsf{x}\phi_a)^2 - \frac{\alpha}{2}\hat{\Phi}(\mathsf{x})(1+\cos\phi_a)\hat{v}_a - \beta\hat{v}_a]. \end{aligned} \tag{12}$$

There is one Goldstone mode in the $\hat{u}_s$ branch, corresponding to the rotation in the XY plane. As α increases, we will again observe the flattening of u_a and v_s bands. We plot an example in the twisted phase in the figure S3 below.

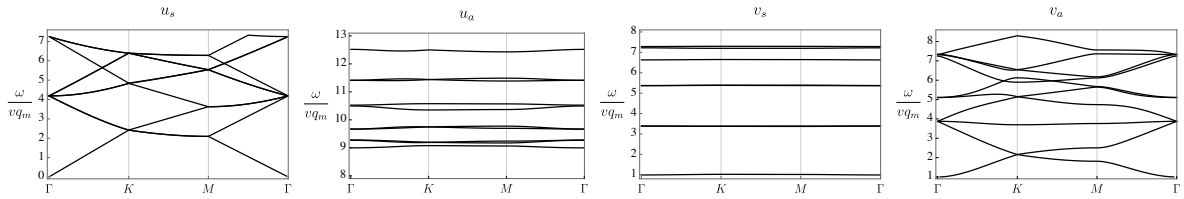

Fig. S3. The ten lowest magnon bands for the four branches at $\alpha = 9$, $\beta = 1$ in the XY-anisotropy case.

B. General perturbative solution of the Euler Lagrange equations. We will encounter the following energy functional and the subsequent partial differential equation in different situations in this work and thus we will first present a general study here. One needs to minimize an energy density functional of the following form:

$$\mathsf{H}_{\mathrm{cl}} = \frac{1}{2}\left|\boldsymbol{\nabla}_\mathsf{x}\phi\right|^2 - \alpha\left(\xi(\mathsf{x}) + \xi_0\right)\cos\phi. \tag{13}$$

$\xi(\mathsf{x})$ is a periodic function defining a triangular lattice, and it has zero mean $\int_{\text{unit cell}} d^2\mathsf{x}\,\xi(\mathsf{x}) = 0$ over one unit cell. One can write a Fourier expansion for $\xi(\mathsf{x})$ in terms of the reciprocal lattice vectors $\hat{\boldsymbol{q}}$ of the above traingular lattice:

$$\xi(\mathsf{x}) = \sum_{\hat{\boldsymbol{q}}\neq 0}\xi_{\hat{\boldsymbol{q}}}\, e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}}, \tag{14}$$

We will be seeking solutions for ϕ that minimize the energy functional above and are periodic with the same period as that given by $\xi(\mathsf{x})$. There is always a trivial solution $\phi = 0$, with an average energy per unit cell equal to $-\xi_0\alpha$. Here we present a perturbative calculation of a nontrivial solution when α and ξ_0 are small; these two parameters are taken to be small with their ratio $\frac{\xi_0}{\alpha}$ kept a constant.

In order to find the function ϕ that minimizes the above energy functional, we will use the following Euler-Lagrange equation:

$$\nabla_{\mathsf{x}}^2 \phi = \alpha \left(\xi(\mathsf{x}) + \xi_0\right) \sin\phi, \tag{15}$$

which should be solved with periodic boundary conditions. Since we are interested in the specific limit of both α and ξ_0 being small, while keeping their ratio $\delta = \frac{\xi_0}{\alpha}$ a constant, we will add a bookkeeping parameter ε to keep track of orders in our perturbation; we will ultimately set $\varepsilon = 1$. The equation thus takes the form:

$$\nabla_{\mathsf{x}}^2 \phi = \varepsilon\ \alpha \left(\xi(\mathsf{x}) + \varepsilon\xi_0\right) \sin\phi. \tag{16}$$

We will find a nontrivial solution as a power series in ε:

$$\phi = \phi^{(0)} + \varepsilon\, \phi^{(1)} + \varepsilon^2\, \phi^{(2)} + \dots \tag{17}$$

To zeroth order in ε, one needs ϕ to be a constant, this constant will be determined in higher orders:

$$\phi^{(0)} = \text{const.}. \tag{18}$$

To first order in ε, the differential equation takes the form:

$$O(\varepsilon): \qquad \sum_{\hat{\boldsymbol{q}}\neq 0} \left(-\left|\hat{\boldsymbol{q}}\right|^2\right) \phi^{(1)}_{\hat{\boldsymbol{q}}} e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}} = \alpha \sin\phi^{(0)} \sum_{\hat{\boldsymbol{q}}\neq 0} \xi_{\hat{\boldsymbol{q}}} e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}}. \tag{19}$$

It could be satisfied if $\phi^{(1)}$ takes the form:

$$\phi^{(1)} = -\alpha \sin\phi^{(0)} \left(\sum_{\hat{\boldsymbol{q}}\neq 0} \frac{1}{\left|\hat{\boldsymbol{q}}\right|^2}\, \xi_{\hat{\boldsymbol{q}}}\ e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}}\right)\ +\ \phi^{(1)[\hat{\boldsymbol{q}}=0]}, \tag{20}$$

where $\phi^{(1)[\hat{\boldsymbol{q}}=0]}$ denotes a constant needed in the first order solution, this constant should also be fixed using higher orders of the equation. The second order in ε of the differential equation now reads:

$$O(\varepsilon^2): \qquad \sum_{\hat{\boldsymbol{q}}\neq 0} \left(-\left|\hat{\boldsymbol{q}}\right|^2\right) \phi^{(2)}_{\hat{\boldsymbol{q}}} e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}} = \alpha \cos\phi^{(0)} \left(\sum_{\hat{\boldsymbol{q}}_1,\hat{\boldsymbol{q}}_2\neq 0} \xi_{\hat{\boldsymbol{q}}_1} \phi^{(1)}_{\hat{\boldsymbol{q}}_2}\ e^{i(\hat{\boldsymbol{q}}_1+\hat{\boldsymbol{q}}_2)\cdot\mathsf{x}}\right) + \alpha\xi_0 \sin\phi^{(0)}. \tag{21}$$

The left hand side of the above equation does not contain a $\hat{\boldsymbol{q}} = 0$ component while the right hand side does:

$$-\,\alpha^2 \cos\phi^{(0)} \sin\phi^{(0)} \sum_{\hat{\boldsymbol{q}}\neq 0} \frac{1}{\left|\hat{\boldsymbol{q}}\right|^2}\ \left|\xi_{\hat{\boldsymbol{q}}}\right|^2 + \alpha\xi_0 \sin\phi^{(0)}. \tag{22}$$

This needs to vanish so that the second order differential equation holds, and this fixes the value of $\phi^{(0)}$:

$$\cos\phi^{(0)} = \frac{\xi_0}{\alpha} \frac{1}{\sum \frac{1}{|\hat{\boldsymbol{q}}|^2}\ \left|\xi_{\hat{\boldsymbol{q}}}\right|^2} = \delta\ \frac{1}{\sum \frac{1}{|\hat{\boldsymbol{q}}|^2}\ \left|\xi_{\hat{\boldsymbol{q}}}\right|^2}. \tag{23}$$

This result shows that for values of δ smaller than $\sum \frac{1}{|\hat{\boldsymbol{q}}|^2}\ \left|\xi_{\hat{\boldsymbol{q}}}\right|^2$, a nontrivial solution could exist. Furthermore, the second order part of ϕ could be found also using Eq. (21):

$$\begin{aligned}\phi^{(2)} = {}& \alpha^2 \sin\phi^{(0)} \cos\phi^{(0)} \left(\sum_{\hat{\boldsymbol{q}}_1\neq -\hat{\boldsymbol{q}}_2} \xi_{\hat{\boldsymbol{q}}_1}\xi_{\hat{\boldsymbol{q}}_2} \frac{e^{i(\hat{\boldsymbol{q}}_1+\hat{\boldsymbol{q}}_2)\cdot\mathsf{x}}}{\left|\hat{\boldsymbol{q}}_1\right|^2\ \left|\hat{\boldsymbol{q}}_1+\hat{\boldsymbol{q}}_2\right|^2}\right) \\ & - \alpha\cos\phi^{(0)} \left(\sum_{\hat{\boldsymbol{q}}\neq 0} \frac{1}{\left|\hat{\boldsymbol{q}}\right|^2}\, \xi_{\hat{\boldsymbol{q}}}\ e^{i\hat{\boldsymbol{q}}\cdot\mathsf{x}}\right)\ \phi^{(1)[\hat{\boldsymbol{q}}=0]}\ +\ \phi^{(2)[\hat{\boldsymbol{q}}=0]},\end{aligned} \tag{24}$$

with the constant $\phi^{(2)[\hat{\boldsymbol{q}}=0]}$ determined again by higher orders of the differential equation.

This procedure can be carried out order by order, we will just state the result for $\phi^{(1)[\hat{\boldsymbol{q}}=0]}$, which could be derived from the $\hat{\boldsymbol{q}} = 0$ component of the ε^3 order of the differential equation:

$$\phi^{(1)[\hat{\boldsymbol{q}}=0]} = \frac{\alpha}{2}\ \frac{\left(1-3\cos^2\phi^{(0)}\right)}{\sin\phi^{(0)}}\ \frac{\sum_{\hat{\boldsymbol{q}}_1,\hat{\boldsymbol{q}}_3,\hat{\boldsymbol{q}}_3} \frac{1}{|\hat{\boldsymbol{q}}_1|^2\ |\hat{\boldsymbol{q}}_2|^2}\ \xi_{\hat{\boldsymbol{q}}_1}\xi_{\hat{\boldsymbol{q}}_2}\xi_{\hat{\boldsymbol{q}}_3}\ \delta_{\hat{\boldsymbol{q}}_1+\hat{\boldsymbol{q}}_2+\hat{\boldsymbol{q}}_3,0}}{\sum_{\hat{\boldsymbol{q}}} \frac{1}{|\hat{\boldsymbol{q}}|^2}\ \left|\xi_{\hat{\boldsymbol{q}}}\right|^2}. \tag{25}$$

Also, the average energy density per unit cell can be found to be:

$$\overline{\mathsf{H}_{\mathrm{cl}}} = -\frac{\alpha^2}{2} \left(\sum_{\hat{\boldsymbol{q}}\neq 0} \frac{1}{\left|\hat{\boldsymbol{q}}\right|^2}\, \left|\xi_{\hat{\boldsymbol{q}}}\right|^2\right) \left(1+\cos^2\phi^{(0)}\right) + O\left(\alpha^3\right), \tag{26}$$

where $O\left(\alpha^3\right)$ denotes any cubic power of α and ξ_0. This should be compared with the trivial solution energy density, i.e. $-\xi_0\alpha$; the twisted solution, when it exists, has lower energy to this order and thus it is the true ground state for $\delta < \sum \frac{1}{|\hat{\boldsymbol{q}}|^2} |\xi_{\hat{\boldsymbol{q}}}|^2$. At $\delta = \sum \frac{1}{|\hat{\boldsymbol{q}}|^2} |\xi_{\hat{\boldsymbol{q}}}|^2$, interestingly, the two solutions coincide and thus this transition is continuous.

Finally we would like to emphasize that the above perturbative expansion works when both α and ξ_0 are small with their ratio $\delta = \frac{\xi_0}{\alpha}$ kept constant; δ could be small or order one but the perturbation breaks down for large δ. Below, we will elaborate on the three cases that the above perturbative calculation has been used in this work.

C. Twisted antiferromagnets. One should consider solving the following Euler-Lagrange equations for a twisted antiferromagnet as discussed in the main text:

$$\nabla_{\mathsf{x}}^2 \phi_s = \beta \cos\phi_a \sin\phi_s, \tag{27}$$

$$\nabla_{\mathsf{x}}^2 \phi_a = \left(\beta \cos\phi_s + \alpha \hat{\Phi}(\mathsf{x})\right) \sin\phi_a, \tag{28}$$

with $\hat{\Phi}(\mathsf{x}) = \sum_{a=1}^{3} \cos(\hat{\boldsymbol{q}}_a \cdot \mathsf{x})$ and $|\hat{\boldsymbol{q}}_a| = 1$. One can find a nontrivial twisted solution (which we call twisted-s in the main text) by setting $\phi_s = 0$ or π; it will be shown below that the $\phi_s = \pi$ solution has lower energy. Starting from the twsited-s solution, increasing β at small α results in a transtion to the collinear phase, while on the other hand for large α, with increasing β, one encounters a transition to the twisted-a phase. We will discuss these two cases separately below.

Transtion from the twisted-s phase to the collinear phase, large angles. At large angles, both α and β are small and we will treat the Euler-Lagrange equations perturbatively. With choosing $\beta = 0$ or π, the equations read:

$$\nabla_{\mathsf{x}}^2 \phi_a = \alpha \left(\hat{\Phi}(\mathsf{x}) \pm \alpha\, \delta\right) \sin\phi_a, \tag{29}$$

where $+$ corresponds to $\phi_s = 0$ and $-$ corresponds to $\phi_s = \pi$, and we will be considering the limit where the ratio $\delta = \frac{\beta}{\alpha^2}$ is kept constant, so that we can use the perturbation series developed above; $\hat{\Phi}(\mathsf{x})$ plays the role of $\xi(\mathsf{x})$, and $\alpha\, \delta$ plays the role of ξ_0. The ϕ_a solution can be found order by order as discussed above:

$$\phi_a = \cos^{-1}\left(\pm\frac{2}{3}\delta\right) - \alpha \sin\phi^{(0)} \left(\hat{\Phi}(\mathsf{x}) - \left[\frac{1}{2} - \cot^2\phi^{(0)}\right]\right) + O(\alpha^2, \beta). \tag{30}$$

The energy density can also be calculated which leads to:

$$\overline{\mathsf{H}_{\mathrm{cl}}} = -\frac{3}{4}\alpha^2\left(1 + \frac{4}{9}\delta^2\right) \quad \pm \frac{1}{6}\alpha^3\delta\left(1 + 4\delta^2\right) \quad + O\left(\alpha^4\right). \tag{31}$$

This result is kept to one higher order than the previous section; it is this higher order which shows that $\phi_s = \pi$ is preferred energetically and so the $-$ sign should be chosen throughout.

Also, it is worthwhile to note that the limit of $\delta \to 0$, corresponds to $\cos\phi_a^{(0)} = 0$, which simply means that $\boldsymbol{N}_1 \cdot \boldsymbol{N}_2 = 0$ to lowest order in α.

Transition from twisted-s phase to the twisted-a phase, small angles. At small angles, where α is large but β is kept still small, one can take the configuration of ϕ_a to be completely determined by satisfying the $-\alpha\hat{\Phi}(\mathsf{x})\cos\phi_a$ term in the Hamiltonian: $\cos\phi_a$ can be taken equal to sign $\left[\hat{\Phi}(\mathsf{x})\right]$. This forms domains of constant ϕ_a, with narrow domain walls between them. On the other hand, ϕ_s should be found using the Euler-Lagrange equations, which reduce to:

$$\nabla_{\mathsf{x}}^2 \phi_s = \beta \cos\phi_a \sin\phi_s. \tag{32}$$

For small β, we can use the perturbation theory developed above, with β, and $\cos\phi_a = \text{sign}\left[\hat{\Phi}(\mathsf{x})\right]$ (remember that ϕ_a is not dynamical in the above equation) playing the roles of α, and $\xi(\mathsf{x}) + \xi_0$ respectively in Eq. (15). One can see that a twisted solution with $\cos\phi_s^{(0)} = \frac{\xi_0}{\sum \frac{1}{|\hat{\boldsymbol{q}}|^2}|\xi_{\hat{\boldsymbol{q}}}|^2} \frac{1}{\beta}$ exists, if β is large enough. It is found numerically that

$$\xi_0 = \frac{1}{A_{\mathrm{u.c.}}} \int_{\text{unit cell}} \text{sign}\left[\hat{\Phi}(\mathsf{x})\right] = -0.21,$$

which means that domains with antiferromagnetic interlayer exchange have larger area than those with ferromagnetic interlayer coupling. Furthermore, one can also find numerically that

$$\sum \frac{1}{|\hat{\boldsymbol{q}}|^2} |\xi_{\hat{\boldsymbol{q}}}|^2 = 0.71.$$

These two values show that a twisted solution for ϕ_s could appear if $\beta > 0.29$; this should correpsond to the β value for which the transition between twisted-s and twisted-a phases occurs at large α, and it is indeed very close, with a few percent error actually, to the value found numerically at large α in the phase diagram presented in the main text.

D. Twisted ferromagnetic CrI_3 bilayer. For the properties of the interlayer exchange parameter in a bilayer CrI_3 system, we will be following the numerical results presented in Ref. (1), where first-principles calculations are carried out: it is shown that the interlayer exchange can vary considerably if the bilayer stacking is altered, and in fact it can change its sign; the pristine CrI_3 bilayer exhibits antiferromagnetic interlayer exchange, but the above statement implies that this can be modified if the stacking is varied. Remarkably in a twisted bilayer, the displacement between the layers is modulated periodically with a unit cell given by the moiré length and so all the different kinds of displaced bilayer stacking are realized. With this in mind, one can use the energy functional discussed in the main text

$$\mathcal{H}_{\rm cl} = \sum_l \left[\frac{\rho}{2} \left(\nabla \boldsymbol{M}_l \right)^2 - d \left(N_l^z \right)^2 \right] - J' \, \Phi \left(\boldsymbol{u}_1(\boldsymbol{x}) - \boldsymbol{u}_2(\boldsymbol{x}) \right) \boldsymbol{M}_1 \cdot \boldsymbol{M}_2, \tag{33}$$

where, as discussed in the main text, the stacking dependence of interlayer exchange, i.e. the function $\Phi(\boldsymbol{u}_1 - \boldsymbol{u}_2)$, could be extracted from some first principle calculations, for example those carried out in Ref. (1).

We have used the plots presented in Ref. (1), to find the Fourier components of the interlayer exchange which is indeed a periodic function of the interlayer displacement. It turns out that unlike the antiferromagnetic case initially studied in the main text, i.e. $\Phi(\boldsymbol{x}) = \sum_{a=1}^{3} \cos(\boldsymbol{q}_a \cdot \boldsymbol{x})$, the present $\Phi \left(\boldsymbol{u}_1(\boldsymbol{x}) - \boldsymbol{u}_2(\boldsymbol{x}) \right)$ function needs several harmonics along with a constant term to be reproduced (see Fig. S4). We will ultimately work with a rescaled Hamiltonian that has a form that is identical to that in the antiferromagnetic case:

$$\mathsf{H}_{\rm cl} = \frac{1}{2} \left(|\boldsymbol{\nabla}_{\mathsf{x}} \phi_s|^2 + |\boldsymbol{\nabla}_{\mathsf{x}} \phi_a|^2 \right) - \left(\alpha \hat{\Phi}(\mathsf{x}) + \beta \cos \phi_{\mathsf{s}} \right) \cos \phi_{\mathsf{a}}. \tag{34}$$

α and β are defined as before and $\hat{\Phi}(\mathsf{x}) = \hat{\Phi}_0 + \sum_{\hat{\boldsymbol{q}} \neq 0} \hat{\Phi}_{\hat{\boldsymbol{q}}} \, e^{i \hat{\boldsymbol{q}} \cdot \mathsf{x}}$, where $\hat{\boldsymbol{q}}$'s are the rescaled moiré reciprocal lattice vectors. We have kept the lowest five harmonics along with the constant term here. Furthermore, $\hat{\Phi}$ is normalized in a way that $\sum_{\hat{\boldsymbol{q}} \neq 0} \frac{1}{|\hat{\boldsymbol{q}}|^2} \left| \hat{\Phi}_{\hat{\boldsymbol{q}}} \right|^2 = 1$. Variation of this energy functional leads to the same set of equations

$$\nabla_{\mathsf{x}}^2 \phi_s = \beta \cos \phi_a \sin \phi_s, \tag{35}$$

$$\nabla_{\mathsf{x}}^2 \phi_a = \left(\beta \cos \phi_s + \alpha \hat{\Phi}(\mathsf{x}) \right) \sin \phi_a, \tag{36}$$

which should be solved to minimize the energy here also. We will only discuss the case of a positive infinitesimal β here, the case of general positive β should be similar to the antiferromagnetic case. The effect of a positive infinitesimal β is to fix a value for ϕ_s, and in this case it turns out that $\phi_s = 0$ is energetically favored; this will be justified below.

The only functionality that needs to be determined now is that of ϕ_a, and the only parameter is α; this time $\hat{\Phi}(\mathsf{x})$ has a nonzero constant term $\hat{\Phi}_0$ as well, and thus at small α the ϕ_a configuration is totally controlled by this constant term; we have derived $\hat{\Phi}_0 = 0.026 > 0$ which means that this constant imposes ferromagetic interlayer exchange, and thus the solution at small α turns out to be $\phi_a = 0$, with an energy density $\overline{\mathsf{H}_{\rm cl}} = -\alpha \hat{\Phi}_0$. One expects this trivial state to give way to a twisted solution with lower energy at some value of α; since $\hat{\Phi}_0$ is small itself, the transition to a twisted phase happens at a small α and thus one can set up a perturbative calculation for the twisted solution of ϕ_a at small α; this perburbative calculation, which is discussed in Sec. B of the SM, yields

$$\phi_a = \phi_a^{(0)} + \alpha \left(- \sin \phi_a^{(0)} \sum_{\hat{\boldsymbol{q}} \neq 0} \frac{\hat{\Phi}_{\hat{\boldsymbol{q}}}}{|\hat{\boldsymbol{q}}|^2} \, e^{i \hat{\boldsymbol{q}} \cdot \mathsf{x}} + \phi_a^{(1)[\hat{\boldsymbol{q}}=0]} \right) \quad + O(\alpha^2, \alpha \, \hat{\Phi}_0), \tag{37}$$

with $\cos \phi_a^{(0)} = \frac{1}{\alpha} \hat{\Phi}_0$. This means that the twisted solution exists for α above $\alpha_0 = \hat{\Phi}_0 = 0.026$ to leading order. The energy density for this state turns out to be $\overline{\mathsf{H}_{\rm cl}} = -\frac{1}{2} \left(\alpha^2 + \hat{\Phi}_0^2 \right)$ to leading order; this shows that indeed a continuous transition to the twisted phase happens at $\alpha = \alpha_0$.

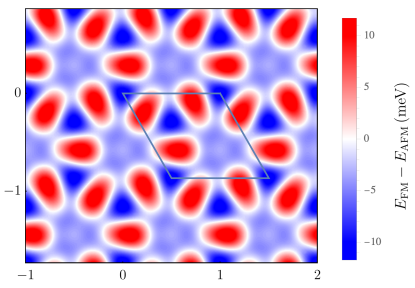

Fig. S4. A reproduced plot of interlayer exchange energy per unit cell of bilayer CrI_3 as a function of the displacement between the two layers. The data is extracted from figures in Ref. (1). The two axes show displacement in the two directions in the units of a real space unit cell length; the interlayer exchange is indeed a periodic function. The blue and red regions show ferromagnetic and antiferromagnetc interlayer exchange.

For very large values of α similar to what happens in the twisted antiferromagnets discussed in the main text, the twisted solution implies that ϕ_a is either 0 or π almost everywhere, so that $\cos\phi_a = \mathrm{sign}[\hat{\Phi}(\mathsf{x})]$ except for narrow domain wall regions where $\hat{\Phi}(\mathsf{x}) = 0$.

Here we can see why $\phi_s = 0$ is chosen for an infinitesimal positive β in two different limits: at small α, the constant term $\hat{\Phi}_0$ is ferromagnetic and thus the energy will decrease by setting $\phi_s = 0$; for large α on the other hand, since one is in the extreme twisted phase, one should note that the area with ferromagnetic interlayer coupling is larger than that with antiferromagnetic coupling, or in other words

$$\frac{1}{A_{\text{u.c.}}} \int_{\text{unit cell}} \mathrm{sign}\left[\hat{\Phi}(\mathsf{x})\right] > 0,$$

and thus $\phi_s = 0$ is again energetically favored. It is worthwhile to mention that this is a coincidence in CrI_3, that both small and large α limits prefer interlayer ferromagnetism; this could well not be the case in other materials in which cases it is reasonable to expect a transition at intermediate α from $\phi_s = 0$ to $\phi_s = \pi$.

References

1. N Sivadas, S Okamoto, X Xu, CJ Fennie, D Xiao, Stacking-dependent magnetism in bilayer cri3. *Nano letters* **18**, 7658–7664 (2018).